

ACRYLIC RESIN-IMPREGNATED BODIES FORMED OF  
EXPANDED GRAPHITE, PROCESS FOR PRODUCING SUCH  
5 BODIES AND SEALING ELEMENTS, FUEL CELL COMPONENTS  
AND HEAT-CONDUCTING ELEMENTS FORMED OF THE BODIES

Background of the Invention:

Field of the Invention:

10 The invention relates to a synthetic resin-impregnated body  
made of expanded or at least partially recompressed expanded  
graphite, a process for producing such a body and sealing  
elements, fuel cell components and heat-conducting elements  
formed of the bodies. In this contest, the phrase "synthetic  
15 resin-impregnated body" is understood to mean a body made of  
expanded graphite which is impregnated by synthetic resin.

Material composites of graphite and plastics are widely used  
in many technical applications. For example, particles of  
20 electrographite are processed with fluoroplastics into highly  
corrosion-resistant components for the construction of  
chemical apparatus, but they are comparatively expensive due  
to the costs of the fluoroplastics and the processing  
technique required. A subject which in terms of content is  
25 even closer to the present application is set out in U.S.  
Patent No. 4,265,952: expanded graphite is mixed with fine

PTFE powder, for example, and subsequently compressed. To that extent, the production technique differs from the impregnating technique described in the present application.

5 Another example of a material composite of graphite and plastic is superficially resin-impregnated foils made of natural graphite, which are predominantly employed in the form of flat seals against particularly aggressive media. Many references to that second example are found in the technical  
10 literature.

Today, thousands of tons of foils made of natural graphite are produced worldwide every year. The process used therefor is described in European Patent Application 0 087 489 A1, U.S.

15 Patent No. 3,404,061 and U.S. Patent No. 3,494,382. The disclosures of those references are incorporated by reference in the present application. To summarize, the following takes place: an intercalating agent such as, for example, concentrated sulfuric acid, acts on natural graphite,

20 preferably platelike or flaky natural graphite, in the presence of an oxidizing agent such as concentrated nitric acid or hydrogen peroxide, for example. That results in graphite intercalation compounds in the graphite flakes or graphite platelets. The flakes are thermally decomposed by  
25 brief heating, for example by introduction into the flame of a gas burner and, as a result of the gas pressure arising in

their interior during that decomposition process, puff up to form loose graphite particles with a wormlike shape. That product is also referred to as "expanded" graphite or as graphite expandate.

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Expanded graphite is extremely plastic and can be readily shaped without the aid of a special binder while being compressed to a greater or lesser degree. Economically, the most important product thus produced is a flexible graphite foil, which can be produced efficiently on calender belts. Such products have typical bulk densities of between 0.7 and 1.3 g/cm<sup>3</sup>. However, other parts having different geometry, for instance individual sealing bodies which, on average, are compressed to a greater degree and have bulk densities of 1.0 to 1.8 g/cm<sup>3</sup>, are also possible. There are also sponge-like parts on average having low bulk density, with values of 0.1 to 1.0 g/cm<sup>3</sup>. All of those bodies with different shapes and different bulk densities have an open pore system. They are referred to hereinbelow as a "primary product".

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Material composites formed of such a primary product and of synthetic resins or plastics materials perform a variety of tasks. Synthetic resins or plastics materials lower the permeability, improve the surface properties, for example the scratch resistance, increase the strength to a small extent, lower the thermal stability of a material composite containing

expanded graphite, and can reduce the electrical conductivity or modify the resistance to media. An expedient technique for the production of the material composites is impregnation.

- 5 According to German Patent DE 32 44 595 C2, corresponding to UK Patent Application GB 2 131 500 A, the sticking action of graphite foils to metal surfaces can be reduced by impregnating the primary product with furan resin in regions close to the surface.

- 10 According to the prior art, the substantial impregnation of shaped bodies made of expanded and partially recompressed graphite is difficult. In order to overcome such difficulties, International Publication No. WO 99/16141 (U.S. Patent No. 6,037,074) teaches that such a body can be
- 15 satisfactorily impregnated when it is interspersed with mineral fibers, which also pass through the surface of the particular bodies. In that way, small channels are formed along those mineral fibers, in which the resin can flow into
- 20 the interior of the bodies during the impregnation. In that reference, a phenolic resin dissolved in acetone, i.e. a solvent-containing thermosetting resin with condensation reactions during the curing, is cited as the impregnating agent.

Another method for achieving good impregnation of bodies made of expanded graphite resides in converting the desired resins through the use of solvents into low-viscosity liquids, whereby the impregnation becomes more complete. In Published Japanese Patent Application JP 1 100 040 A2, the thermosetting resins cited are based on phenols, epoxides, polyimides, melamines, polyesters and furans, which are used in a mixture solution with polyvinylbutyral.

Published Japanese Patent Application JP 1 308 872 A2 describes the solution to other problems. A material composite formed of a glass fiber nonwoven fabric and an expanded graphite foil is produced in order to thus strengthen the latter and overall obtain a liquid-tight material. That is achieved by impregnating with epoxy resin. The resin penetrates the nonwoven fabric and at the same time also penetrates into the surface, i.e. partially into the foil. During subsequent curing, the supporting part of the composite material is formed from the impregnated nonwoven fabric and is then also sealed at the surface.

The impregnation of expanded graphite foil with phenolic resin or epoxy resin, set out in Published Japanese Patent Application JP 60 24 2041 A2 (German patent DE 35 12 867 C2), serves similar purposes, namely to improve strength and gas-tightness. The special feature in that case lies in a

degassing process for the liquid resins and the foil present therein which is repeated a number of times, presumably with the aim of improving the quality of the impregnation.

- 5 German Published, Non-Prosecuted Patent Application DE 43 32 346 A1 describes the impregnation of the expanded graphite foils for the purpose of improving adhesion to elastomer layers lying thereon. The viscosity of the epoxy resins used in that case is 2100 to 2400 mPa.s.

10 Published Japanese Patent Application JP 11 35 4136 A2 entitled "Fuel Cell, Separator for Fuel Cell, and Manufacture Therefor" describes the production of expanded graphite in sheet-like form. That partially recompressed expanded  
15 graphite is subsequently comminuted (pulverized) and then mixed selectively with resins, solvent-free epoxy resin, solid epoxy resin, melamine resin, acrylic resin, phenolic resin, polyamide resin, and the like. The mixture is subsequently shaped. As will be shown below, that technique differs from  
20 the bodies according to the invention which have an entirely different structure in that the resins are mixed into an expanded graphite granulate.

International Publication No. WO 98/09926 describes a graphite  
25 foil which is coated with a plastic on at least one side. That is accomplished firstly with an aqueous solution of an

acrylic resin which is applied to the surface, remains there, but also penetrates into regions of the foil close to the surface, and is then dried in.

5 The prior art set out above discloses various synthetic resin-containing bodies produced by using expanded graphite and processes for their production. That it is difficult to produce high-quality, synthetic resin-containing graphite bodies from recompressed, expanded graphite is easy to see.

10 All of the described processes have disadvantages, some of which are serious: if resins that are diluted by solvents and thus have lower viscosity are used during the impregnation, it is true that the impregnation is easier. However, the vapors from the, in most cases, readily volatile solvents cause

15 serious problems during the impregnation itself, especially during subsequent process steps. In particular, as a result of the fact that they escape during the curing of the resins, they leave behind fine channels which raise the permeability of the bodies being produced. If an increased permeability

20 can neither be tolerated nor is desired, there is furthermore a general problem: if the curing is not performed very slowly, i.e. is time-consuming, blisters and cracks are formed in the bodies, which lower their quality considerably. The same

25 applies to resin systems which release gases from condensation reactions during the curing.

As a result of the fact that solvents or other gases and vapors escape, a residual porosity arises in the bodies. Attempts are now frequently made to eliminate the residual porosity by one or more additional impregnating operations.

5 The attendant increase in expenditure is clear and the success is really limited. In addition, solvent-containing resins always above all require measures to allow their safe handling and the harmless removal or recovery of the solvents, which increases the expenditure even further. However, the solution  
10 of the problem through the addition of fibers penetrating the surfaces of the body may improve the impregnating properties of the body but does not eliminate the problems outlined for the use of solvent-containing resins releasing vapors or gases. In addition, one always has a product containing  
15 certain fibers, which is more expensive to produce.

The problems with solvents present in the resin systems which have been discussed also apply to aqueous resins, for example according to International Publication No. WO 98/09926.

20 Furthermore, according to that publication, a graphite foil is provided with a resin system which results in the formation of a coat of plastics material at the surface for the purpose of reinforcement. During the application of that resin, it also penetrates into the surface of the foil. The plastics  
25 material coating on one hand has the effect that a second



coating with improved adhesion can be applied and on the other hand has the effect of electrical insulation.

Both aspects, i.e. the resin system dissolved in water and the electrical insulation at the surface of the body, are regarded as disadvantageous for the use of the bodies according to the present application.

#### Summary of the Invention:

It is accordingly an object of the invention to provide acrylic resin-impregnated bodies formed of expanded or at least partially recompressed expanded graphite having a liquid-accessible pore system which is completely or partially filled with an uncured or partially or completely cured synthetic resin, a process for producing such bodies and sealing elements, fuel cell components and heat-conducting elements formed of the bodies, which overcome the hereinafore-mentioned disadvantages of the heretofore-known products and processes of this general type and in which the body does not contain any defects such as blisters or cracks that may be caused by reactions of the synthetic resin during the curing, the body is producible with comparatively little expenditure and the body is corrosion-resistant, electrically and thermally conductive and is from liquid-permeable to gas-tight, depending on the degree of compression.

With the foregoing and other objects in view there is provided, in accordance with the invention, a synthetic resin-impregnated body, comprising expanded or at least partially recompressed expanded graphite. The graphite contains either  
5 at least one solvent-free, low-viscosity, storage-stable, polymerizable acrylic resin system or polymers obtained by curing the at least one resin system.

Therefore, the object of the present invention is achieved  
10 with a body of the type mentioned at the outset by subjecting the primary product or the body obtained from the impregnated primary product to at least partial compression containing  
15 either solvent-free, low-viscosity, storage-stable acrylic resin systems or cured acrylic resin systems. The resin systems enter into the body by impregnating the primary product with solvent-free, low-viscosity, storage-stable and polymerizable acrylic resin systems.

In order to eliminate the aforementioned disadvantages of  
20 solvent-containing resin systems and to achieve the advantages of resin systems of low viscosity, the following special solvent-free resin systems were employed according to the invention presented herein:

25 The main component is triethyleneglycol dimethacrylate and the initiator systems come from the azo initiators group.

Examples are 2,2'-dimethyl-2,2'-azodipropiononitrile and/or 1,1'-azobis(1-cyclohexanecarbonitrile) and/or azoisobutyric acid dinitrile. A possible selection of the proportions of the individual components in the overall mixture is mentioned in the examples.

The low viscosities at processing temperature of the resin systems ensure good and efficient impregnation of the primary product and the polyadditions which take place during the curing do not give rise to any low-molecular-weight cleavage products, which could cause blistering or even cracks in the body. The testing of the resin systems is described in more detail in the examples.

At room temperature, the specified mixture has a viscosity of between 10 and 20 mPa·s which is markedly below that of solvent-free, low-viscosity, storage-stable and polymerizable resin systems from the group of isocyanates and their co-reactants and/or epoxides. The main component of the acrylic resin system can be characterized from the development of the viscosities over time in a unit [mPa·s] at room temperature, as follows: fresh mixture  $\approx$  13, after eight days  $\approx$  13 and after 48 days  $\approx$  14.

The small rate of the changes of the viscosity of the resin at room temperature and over a period of several weeks is demonstrated through the use of these viscosity measurements. That small rate of the changes will be referred to hereinafter by the term "high storage stability".

The expanded graphite used to produce the primary product is formed of fanned-out, wormlike structures, in which very fine graphite platelets are joined together in the form of a defective accordion bellows. During the compression of the primary product, these platelets slide in and over one another. They become interlocked and thus come into contact again so as to no longer be able to be released without destruction. This gives rise in the primary product to a porous graphite framework or network which has good electrical as well as good thermal conductivity due to the good contacts between the graphite platelets. Since these properties are based on the framework function of the graphite in the primary product, they are not adversely affected by the impregnation with synthetic resin. They can even be further improved during a subsequent compression of the primary product impregnated with resin.

The primary product is permeated throughout by open pores which are interconnected in a variety of ways. As a result of this network of interconnected pores, the synthetic resin

penetrates into the primary-product body during the impregnation and may even completely fill it under suitable conditions. The network of pores then becomes a network of synthetic resin. Both networks, the graphite network and the pore/synthetic resin network, in combination result in the outstanding properties of the end products thus produced. By adjusting them in a specific manner, it is also possible to control the level of properties of the end products. For example, on one hand, a primary-product body which has undergone little precompression and is thus highly porous has a lower electrical and thermal conductivity and a lower degree of anisotropy than a more highly compressed primary-product body. On the other hand, it can take-up more synthetic resin and has modified strength properties. This situation is reversed with greatly compressed primary-product bodies. After the impregnation and curing of the synthetic resin, they yield products with improved electrical and thermal conductivity, as well as good mechanical strengths. All of the bodies according to the invention which are described herein are highly impermeable to liquids and gases when their pore network has been completely filled with synthetic resin.

All of the known methods, such as those described in DE 35 12 867, for example, can be used for the impregnation of the primary-product bodies. It is preferable, however, to use immersion methods, in particular immersion methods with prior

evacuation of the vessel containing the primary-product body and flooding of the evacuated vessel with the synthetic resin.

Where appropriate, the vessel is also subjected to a gas pressure after it has been flooded with the synthetic resin.

- 5 If the primary-product body is to be merely impregnated close to the surface or is to be partially impregnated, the impregnating period is shortened or the surfaces from which the impregnation is to start are suitably coated or sprayed with synthetic resin or the body is only partially immersed.

10 Following this treatment, the excess resin is removed from the surface.

15 An essential aspect of the present invention is efficient and damage-free impregnation and curing. The rapid blister-free and crack-free curing which is possible by virtue of the polyaddition reactions has been discussed above. Efficient impregnation depends essentially on the viscosity of the resin system. The present acrylic resin system has a very low viscosity at less than 20 mPa·s, which is why the impregnating  
20 success is very high.

The primary product can take-up an amount of up to 100% of its own weight of resin, depending on the degree of compression of the primary product and the open pore volume conditional  
25 thereon. If, however, a high electrical conductivity of the end product is desired, it is expedient to start with a

primary-product body which has undergone greater precompression, has a lower open pore volume and can then take-up, for example, only 20% by weight of resin based on its own weight. After the curing of the resin, such a body can be

5 highly impermeable to liquids and gases, as is seen in Table 2, and has good strength properties.

The kinetics of the curing reaction are extremely temperature-dependent with the acrylic resin systems that are used.

10 Whereas at room temperature virtually no curing reaction takes place, at higher temperatures and when the azo initiators become effective, it starts suddenly. Virtually no viscous transition state of the resin systems can be observed. The resin systems cure fully all of a sudden. The curing times of

15 the acrylic resin systems fall in proportion to the rise in the temperature. Examples are:

Temperature	60°C	80°C	100°C	150°C
Curing time	days	35 minutes	10 minutes	1 minute

If larger series of components or bodies are to be produced by

20 using the techniques described above, it will be desired to efficiently combine a number of process steps. This is possible particularly with the shaping of impregnated primary-product bodies with simultaneous curing. For this purpose, the impregnated primary product, which is generally in the

form of a semifinished product or blank, is expediently put into a mold which is already hot and the mold is closed. The semifinished product thereby takes on the desired geometry, is simultaneously thoroughly heated and cures completely.

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A relatively wide variety of graphites based on synthetic production and natural occurrence exists, both types being mentioned in U.S. Patent No. 3,404,061. Only natural graphite will be discussed hereinbelow, the graphite which is present as raw material in the bodies described herein.

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Natural graphite is obtained by mining and is separated from the gangue rock with considerable effort. Nevertheless, very small amounts of rock also remain, attached to the natural graphite flakes or having intergrown into the flakes. Those "foreign constituents" are characteristic of every source of natural graphite and can also be specified as an ash value. A method for determining such ash values is described in DIN (German Industrial Standard) 51 903 under the title "Testing of Carbon Materials - Determination of the Ash Value".

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In view of the end uses of the synthetic resin-containing graphite bodies according to the invention, the ash values and ash composition of the graphite that is present are quite important. If such bodies are employed, for example, as inherently corrosion-resistant seals in installations



subjected to corrosive media, certain ash constituents together with the corrosive medium may result in pitting in the corrosion-resistant seals adjoining flanges or bushes of stuffing-box packings and eventually lead to the failure of the sealed joint.

10 Another example of a possible adverse effect of too high an ash value or an unfavorable ash composition of the graphite in a synthetic resin-containing body according to the invention is found in fuel cell technology. Thus, for example, bipolar plates of proton exchange membrane fuel cells can be produced from the material according to the invention. If such a plate has too high an ash content, some of the harmful ash constituents may be released from the plate during the operation of the fuel cell and poison the sensitive catalysts located close to the surfaces of the bipolar plate, resulting in a premature loss of power of the cell.

20 Due to the potential adverse effects of an excessively high ash content, the ash content of the graphite used to produce the bodies according to the invention is 4 per cent by weight and less, preferably less than 2 per cent by weight and in special cases no more than 0.15 per cent by weight.

25 It may be convenient to strengthen the body according to the invention with fillers. The selection of the fillers has to

be matched to the application (e.g. fuel cell). Fillers may be electrically conductive materials closely related to expanded natural graphite, such as, for example, materials from the group consisting of naturally occurring flake  
5 graphites, synthetically produced electrographites, carbon blacks or carbons, and graphite or carbon fibers. Furthermore, use may be made of silicon carbide in granular or fibrous form or else electrically non-conductive ceramic or mineral fillers in granular, platelike or fibrous form, such  
10 as silicates, carbonates, sulfates, oxides, glasses or selected mixtures thereof.

The bodies according to the invention can be used wherever electrically and thermally conductive components having low  
15 weight together with good corrosion resistance are required. Further properties which are essential for various applications are low ash values and relatively high impermeability. The bodies according to the invention are used in particular for components of fuel cells, for seals and  
20 for heat-conducting elements, for example for conducting away excess heat from integrated circuits.

Other features which are considered as characteristic for the invention are set forth in the appended claims.

Although the invention is described herein as embodied in acrylic resin-impregnated bodies formed of expanded graphite, a process for producing such bodies and sealing elements, fuel cell components and heat-conducting elements formed of the bodies, it is nevertheless not intended to be limited to the details given, since various modifications and structural changes may be made therein without departing from the spirit of the invention and within the scope and range of equivalents of the claims.

The construction and method of operation of the invention, however, together with additional objects and advantages thereof will be best understood from the following description of specific embodiments when read in connection with the accompanying examples. In addition, methods for obtaining data on electrical properties and gas-tightness for the examples are dealt with now.

In order to determine gas-tightness, a resin-impregnated graphite body was pressed as a separating plate (test specimen) between two chambers of a testing apparatus. A constantly maintained helium gas pressure of 2 bar absolute prevailed in a first chamber. A metal grid which mechanically supported the test specimen was disposed in a second chamber. In addition, this chamber was connected at ambient pressure to

a liquid-filled burette such as is used, for example, in the leakage measurement of flat seals according to DIN 3535.

5 The helium gas emerging from the first chamber and diffusing through the test specimen was collected in the second chamber and measured by displacement of the liquid in the burette. It was thus possible to determine the volume of the helium gas which diffused through the sample per unit of time. A leakage rate was ascertained which is specified by the unit  $\text{mg}/(\text{m}^2 \cdot \text{s})$  by taking the helium density and the testing area into account.

10 The material composite of partially recompressed expanded graphite and synthetic resin has anisotropic properties, i.e. 15 the individual graphite platelets of the expanded graphite have a preferred orientation due to the production technique. For example, the electrical resistance parallel to this preferred orientation is low and perpendicularly thereto it is higher. In the present case, the cured shaped bodies 20 according to the invention were characterized comparatively by measuring the electrical resistance perpendicularly to the preferred orientation of the graphite layers. For this purpose, the body was clamped between two gold-plated electrodes having a diameter of 50 mm, with defined and in 25 each case identical surface pressure. The electrical resistances R established with the aid of a device (Resistomat

2318) from the firm Burster (Gernsbach, Germany) are specified by the magnitude [mΩ] hereinbelow.

### Example 1

- 5 The following primary-product plates were impregnated at room temperature by immersion:

	Type of primary-product plate	Thickness [mm]	Bulk density [g/cm <sup>3</sup> ]	Ash value [% by weight]
Example 1a	F02510C	0.25	1.0	< 2.0
Example 1b	L10010C	1.0	1.0	< 2.0
Example 1c	L40005Z	4.0	0.5	< 0.15

Table 1:

Primary-product plates made of partially recompressed expanded graphite used for impregnation with an acrylic resin system

The resin system which was used had the following composition:

99.2% of triethyleneglycol dimethacrylate

(methacrylic acid ester)

0.3% of 2,2'-dimethyl-2,2'-azodipropiononitrile

0.5% of 1,1'-azobis(1-cyclohexanecarbonitrile)

The methacrylic acid ester came from the firm Röhm GmbH

(Darmstadt, Germany) and had the trade name PLEX 6918-O. The

two other components of the resin system had the function of

an initiator. 2,2'-Dimethyl-2,2'-azodipropiononitrile came from the firm Pergan GmbH (Bocholt, Germany) and had the trade name Peroxan AZDN. 1,1'-Azobis(1-cyclohexanecarbonitrile) came from the firm Wako Chemicals GmbH (Neuss, Germany) and bore the designation V40. The viscosity of the resin system was in the range from 10 - 15 mPa.s at room temperature.

The primary-product plates were completely immersed in the resin bath and after one, five and nine hours they were removed from the immersion bath and the resin adhering to the surface was wiped off. The plates were subsequently put into a circulating-air oven at 100°C and cured for 30 min. The impregnated primary-product plates showed no blisters or cracks at all despite this shock curing. The values of the resin content, volume resistance R and helium permeability  $\lambda$  determined on the plates are summarized in Table 2 and compared with the values for non-impregnated plates.

Type of primary-product plate	Impregnating period 1 h				Impregnating period 5 h				Impregnating period 9 h			
	Resin content [%]	R [mQ]	$\lambda$ [mg/(m <sup>2</sup> ·s)]		Resin content [%]	R [mQ]	$\lambda$ [mg/(m <sup>2</sup> ·s)]		Resin content [%]	R [mQ]	$\lambda$ [mg/(m <sup>2</sup> ·s)]	
F02510C	23.8	0.10	0.010		33.3	0.19	-		36.1	0.37	< 0.001	
L10010C	10.2	0.35	0.016		16.3	0.48	0.005		19.9	0.43	< 0.001	
L40005C	24.8	0.77	-		37.4	0.76	-		39.3	0.77	-	

without impregnation

Type of primary-product plate	without impregnation			
	Resin content [%]	R [mQ]	$\lambda$ [mg/(m <sup>2</sup> ·s)]	
F02510C	0	< 0.1	1.5	
L10010C	0	0.28	0.323	
L40005C	0	0.43	-	

Table 2:

Comparison of primary-product plates (varying thickness and bulk density) impregnated with an acrylic resin system with non-impregnated primary-product plates (of likewise varying thickness and bulk density). The volume resistances R and the helium permeabilities  $\lambda$  after various impregnating periods are compared.

As is evident from Table 2, the resin content of the composite materials is greatly dependent on the bulk density of the primary product, its geometry (plate thickness) and the impregnating time. The volume resistance of the impregnated plates rises comparatively little with increasing resin content, since the electron conduction is borne by the existing graphite network. The helium permeability of the plates is drastically reduced by the impregnating treatment. Depending on the resin content of the plate, the permeability falls by more than 2 powers of ten as compared with corresponding primary-product plates without impregnation.

#### Example 2

The resin system that was used was the same as the resin system in Example 1. The primary product had a thickness of 2.7 mm and a density of  $0.65 \text{ g/cm}^3$  and the ash value of the graphite was less than 0.15% by weight. After an impregnating period of one hour at room temperature, the now impregnated plate was taken out of the resin bath and weighed after the resin adhering to the surface had been wiped off. The proportion of resin which was determined was 20% by weight. The impregnated plate was placed in a pressing die preheated to  $150^\circ\text{C}$ . The die, which was furnished with an anti-stick coating, was closed and the impregnated graphite was pressed into the mold, in the course of which a further compression of the composite material took place. After five minutes under



the effect of pressing force and temperature, the die was opened and the cured shaped body was removed. The shaped body was free from cracks and blisters and the surface showed no resin film visible to the eye.

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Besides these above-mentioned examples, a multiplicity of further bodies and procedures can be realized according to the teaching of the present invention. Accordingly, the invention is not restricted to the embodiments provided in the examples.

1.0 Variants which are not described but which a person skilled in the art could produce due to the information offered by this disclosure are therefore also to be included in the present patent application.